

Measure of Quantum Macroscopicity for Arbitrary Spin Systems and Quantum Phase Transition as a Genuine Macroscopic Quantum Phenomenon

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We propose a general and computable measure of quantum macroscopicity for arbitrary spin states by quantifying interference fringes in phase space. It effectively discriminates genuine macroscopic quantum effects from mere accumulations of microscopic quantum effects in large systems. The measure is applied to several examples and it is found to be consistent with some previous proposals. In particular, we investigate many-body spin systems undergoing the quantum phase transition (QPT) and the QPT turns out to be a genuine macroscopic quantum phenomenon. Our result suggests that a macroscopic quantum superposition of an extremely large size may appear during the QPT.

Introduction.— As illustrated in Schrödinger’s famous cat paradox [1], quantum mechanics does not preclude the possibility of a macroscopic object being in a quantum superposition. There has been interesting progress in generating macroscopic superpositions using atomic/molecular systems [2, 3], superconducting circuits [4, 5], and optical setups [6–8]. On the other hand, how to sharply define and quantify “macroscopic quantumness” or “quantum macroscopicity” is a slippery issue [9]. Among various ideas, a remarkable notion introduced by Leggett [10] is that genuine macroscopic quantum effects should bring about increasingly more quantum effects than mere accumulation of microscopic quantum effects. For example, Debye’s T^3 law, Bose-Einstein condensates, superconductivity and superfluidity are not considered as genuine macroscopic quantum effects in this category [10, 11].

There are a number of proposals for quantification of macroscopic quantum effects and related discussions [9–29]. While most of the proposals are limited to specific forms of states, recently, more general measures were suggested for arbitrary bosonic systems based on interference fringes in phase space [20] and for arbitrary spin systems in terms of the non-classical parameter estimation represented by the quantum Fisher information (QFI) [21]. However, it is difficult to use the QFI based measure for large spin systems because of its computational complexity due to the requirement of density matrix diagonalization [21].

In this paper, we propose a general and computable measure of quantum macroscopicity that can be applied to arbitrary spin systems. It is based on interference fringes in phase space so that it has a strong conceptual connection with the one for bosonic systems [20]. It is also directly related to the maximum purity decay rate under the Lindblad-type decoherence model. It has a similar mathematical structure as the QFI based measure [21] and the two measures become identical for pure states. However, unlike the QFI based measure, our measure does not require calculations with heavy computational complexities because the density matrix diagonal-

ization is not needed. Our measure is thus closely related to both the previous measures [20, 21] but with its own merit; it can be readily applied to large spin systems to effectively identify genuine macroscopic quantum effects. In search of genuine macroscopic quantum phenomena, we investigate many-body spin states undergoing quantum phase transition (QPT). In contrast to examples such as Debye’s T^3 law, superconductivity and superfluidity [10], the QPT turns out to be a genuine macroscopic quantum phenomenon where the entire effect cannot be explained by accumulative effects of constituting elements.

Quantum macroscopicity for arbitrary spin systems.— It was shown that quantum macroscopicity of arbitrary harmonic oscillator states can be quantified based on its Wigner function structure in the phase space [20]. It is a separate problem to find whether this type of approach is possible for spin systems. The Wigner (or Stratonovich-Weyl) distribution for a spin- S particle is represented by [1]

$$W(\mathbf{n}) = \sqrt{\frac{4\pi}{2S+1}} \sum_{L=0}^{2S} \sum_{M=-L}^L \chi_{L,M}^{(S)} Y_{L,M}(\mathbf{n}), \quad (1)$$

where $Y_{L,M}(\mathbf{n})$ denotes spherical harmonics with a three dimensional unit vector $\mathbf{n}=(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ and $\chi_{L,M}^{(S)} = \text{Tr}[\hat{T}_{L,M}^{(S)\dagger}\rho]$ is the characteristic function with is the irreducible tensor operator $\hat{T}_{L,M}^{(S)}$. The matrix elements of the irreducible tensor operator is defined as $\langle S, m' | \hat{T}_{L,M}^{(S)} | S, m \rangle = \sqrt{(2L+1)/(2S+1)} C_{S,m;L,M}^{S,m'}$ with Clebsch-Gordan coefficients $C_{S,m;L,M}^{S,m'}$ and an eigenstate $|S, m\rangle$ of the z -component spin operator \hat{S}_z with its eigenvalue m . Here we work with a natural unit of $\hbar = 1$.

In order to define a measure of quantum macroscopicity for an arbitrary spin system, we attempt to simultaneously quantify both (i) the distinctness between the component states of a superposition state and (ii) the degree of genuine quantum coherence between those component states against their classical mixture. An important observation is that these properties are closely related to

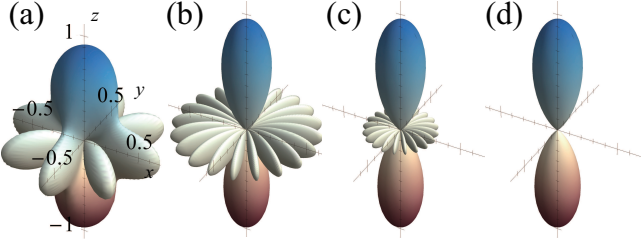


FIG. 1: (Color online) Wigner distributions of $(|S, S\rangle + |S, -S\rangle)/\sqrt{2}$ for (a) $S = 2$ and (b) $S = 5$. Each distribution consists of two peaks along the $\pm z$ direction and interference fringes around the z -axis. When the pure superposition with $S = 5$ becomes a mixed state, $\{|5, 5\rangle\langle 5, 5| + |5, -5\rangle\langle 5, -5| + \gamma(|5, 5\rangle\langle 5, -5| + |5, -5\rangle\langle 5, 5|)\}/2$, the interference fringes are obviously reduced for (c) $\gamma = 1/2$ and they completely disappear for (d) $\gamma = 0$.

two quantities: the *frequency* and the *magnitude* of the interference fringes of the Wigner function. Remarkably, this is consistent with the case of harmonic oscillator systems that leads to the definition of quantum macroscopicity for bosonic states previously investigated in Ref. [20].

To clarify, let us first consider an example of a superposition of two spin- S components of opposite signs, $(|S, S\rangle + |S, -S\rangle)/\sqrt{2}$. We plot its Wigner distribution in a three-dimensional phase space for $S = 2$ in Fig. 1(a) and for $S = 5$ in Fig. 1(b). Once the Wigner distribution is given, the expectation value of an arbitrary spin operator \hat{J} can be calculated by the overlap relation $\langle \hat{J} \rangle = (2S + 1)/(4\pi) \int d\Omega W_J(\mathbf{n})W(\mathbf{n})$, where $d\Omega = \sin\theta d\theta d\phi$ and $W_J(\mathbf{n})$ is the Wigner representation of operator \hat{J} defined in the same manner as the Wigner distribution by replacing the density operator ρ by \hat{J} . As shown in Figs. 1(a) and 1(b), the superposition of a larger value of S shows more frequent periodic patterns around the z -axis. These are the interference fringes of the superposition between the two component states, $|S, S\rangle$ and $|S, -S\rangle$, with perfect quantum coherence. When it undergoes dephasing, it becomes a mixed state as $(|S, S\rangle\langle S, S| + \gamma|S, S\rangle\langle S, -S| + \gamma|S, -S\rangle\langle S, S| + |S, -S\rangle\langle S, -S|)/2$ with $0 \leq \gamma < 1$. The magnitude of the fringes then becomes smaller as shown in Fig. 1(c) and it completely disappears when the state is fully decohered (i.e. $\gamma = 0$) as in Fig. 1(d).

The spherical harmonics $Y_{L,M}(\mathbf{n})$ in Eq. (1) form a complete orthonormal basis to describe an arbitrary spin system. The periodic interference fringes in Fig. 1 are attributed to these spherical harmonics and the ϕ -dependence of $Y_{L,M}(\mathbf{n})$ appears only in the form of $\exp[iM\phi]$. It means that M is the frequency of the interference fringes due to $Y_{L,M}(\mathbf{n})$. We then notice from in Eq. (1) that the complex amplitude of a certain frequency component for M , i.e. $Y_{L,M}(\mathbf{n})$, is $\chi_{L,M}^{(S)}$. Since the two essential elements of quantum macroscopicity are identified, we can define its measure to be proportional to $\sum (\text{frequency}) \times (\text{magnitude})$. Considering the normalization factor, we can straightforwardly attempt a simple

definition as

$$\begin{aligned} \mathcal{I}_z &= \frac{1}{2S} \frac{1}{\mathcal{P}} \sum_{L=0}^{2S} \sum_{M=-L}^L M^2 \left| \chi_{L,M}^{(S)} \right|^2 \\ &= \frac{1}{2S\mathcal{P}} \frac{2S+1}{4\pi} \int d\Omega W(\mathbf{n}) \hat{L}_z^2 W(\mathbf{n}). \end{aligned} \quad (2)$$

where $\mathcal{P}(\rho) = \sum_{L',M'} |\chi_{L',M'}^{(S)}|^2 = \text{Tr}[\rho^2]$ corresponds to the purity of quantum state and $\hat{L}_z = i\partial/\partial\phi$ (see Supplementary Material for the proof). The scaling factor $(2S)^{-1}$ is introduced to make $\mathcal{I}_z = S$ for $\gamma = 1$.

However, the preliminary definition in Eq. (2) is appropriate only for a single spin superposition where the component states are spin- z eigenstates. In order to generalize it to arbitrary directions and arbitrary number of spin systems, we use the angular momentum operator with an arbitrary direction: $\hat{L}_\alpha = \alpha_x \hat{L}_x + \alpha_y \hat{L}_y + \alpha_z \hat{L}_z$, where $\hat{L}_x = i \sin\phi \partial_\theta + i \cot\theta \cos\phi \partial_\phi$, $\hat{L}_y = -i \cos\phi \partial_\theta + i \cot\theta \sin\phi \partial_\phi$ and $\alpha = (\alpha_x, \alpha_y, \alpha_z)$ is a unit vector with a condition $\|\alpha\|^2 = \alpha_x^2 + \alpha_y^2 + \alpha_z^2 = 1$. The multi-spin extension of the definition is

$$\mathcal{I}_{\{\alpha^{(i)}\}} = \frac{1}{2NS\mathcal{P}} \left(\frac{2S+1}{4\pi} \right)^N \int d\Omega W(\{\mathbf{n}_i\}) \hat{L}_{\{\alpha^{(i)}\}}^2 W(\{\mathbf{n}_i\}) \quad (3)$$

where $d\Omega = d\Omega_1 d\Omega_2 \cdots d\Omega_N$ and $\{\mathbf{n}_i\}$ denotes a set of unit vectors $\{\mathbf{n}_1, \dots, \mathbf{n}_N\}$ and $\hat{L}_{\{\alpha^{(i)}\}} = \sum_i \hat{L}_{\alpha^{(i)}}$ is the operator used to capture interference patterns of the total system with many spins, where each local operator is oriented in the direction of $\alpha^{(i)}$. The additional factor N^{-1} was introduced in order to eliminate accumulative microscopic quantum effects according to the size of the system. We then need to find the optimal directions of $\alpha^{(i)}$ for each local i -th spin, which maximizes $\mathcal{I}_{\{\alpha^{(i)}\}}$. For the examples illustrated in Fig. 1, the optimal direction of α is analytically identified to be the z -direction. Now, we have a formal definition of the degree of quantum macroscopicity for a system composed of an arbitrary number of spin- S particles as

$$\mathcal{I}(\rho) = \max_{\{\alpha^{(i)}\}} \mathcal{I}_{\{\alpha^{(i)}\}}. \quad (4)$$

The upper bound of the measure is found to be $\mathcal{I}(\rho) = NS$ for a given particle number N (see the Supplementary Material for the proof). For example, the N -party spin- S Greenberger-Horne-Zeilinger (GHZ) state, $(|S, S\rangle^{\otimes N} + |S, -S\rangle^{\otimes N})/\sqrt{2}$, has the maximum value $\mathcal{I}(\rho) = NS$, where the optimal direction of $\alpha^{(i)}$ is the z -direction regardless of i . On the other hand, $\mathcal{I}(\rho) = S$ for $[(|S, S\rangle + |S, -S\rangle)/\sqrt{2}]^{\otimes N}$ regardless of the number of particles N ; accumulations of microscopic quantum effects do not increase the value of $\mathcal{I}(\rho)$. A spin-1/2 GHZ state of N particles, $(|\uparrow\rangle^{\otimes N} + |\downarrow\rangle^{\otimes N})/\sqrt{2}$, has the same value of $\mathcal{I}(\rho) = N/2$ with a superposition of a single spin- $N/2$ particle, $(|N/2, N/2\rangle + |N/2, -N/2\rangle)/\sqrt{2}$.

We find that $\mathcal{I}(\rho)$ is directly related to a standard decoherence model (for details, see the Supplementary Material) that leads to a more refined expression as

$$\begin{aligned}\mathcal{I}(\rho) &= \frac{1}{NS\mathcal{P}} \max_A \{-\text{Tr}[\rho\mathcal{L}(\rho)]\} = \frac{1}{2NS} \max_A \left\{ -\frac{\dot{\mathcal{P}}}{\mathcal{P}} \right\} \\ &= \max_A \frac{\text{Tr}[\rho^2 A^2 - \rho A \rho A]}{NS\text{Tr}[\rho^2]}\end{aligned}\quad (5)$$

with the Lindblad type of decoherence channel

$$\mathcal{L}(\rho) = \frac{d\rho}{d\tau} = A\rho A^\dagger - \frac{1}{2}(A^\dagger A\rho + \rho A^\dagger A) \quad (6)$$

where $A = \sum_{j=1}^N A^{(j)}$ with $A^{(j)} = \alpha^{(j)} \cdot \hat{\mathbf{S}}^{(j)}$ is the spin operator of the j th particle with $\|\alpha^{(j)}\| = 1$ and τ (decay rate) \times (time) is dimensionless time. Here we find that $\mathcal{I}(\rho)$ may be understood as “the maximum purity decay rate to the purity” of a given state.

If a quantum state is pure, $\mathcal{I}(\rho)$ is reduced to the (normalized) variance of the total spin operator as $\mathcal{I} = \max_A \mathcal{V}(A)/(NS)$, where $\mathcal{V}(A) = \langle A^2 \rangle - \langle A \rangle^2$. Since a macroscopic quantum superposition has well separate component states of the outcome spectrum, it is in agreement with our natural expectation. Of course, the variance itself does not allow one to discriminate between a genuine superposition and a statistical mixture.

Hereafter, we focus on quantum macroscopicity of multi-qubit states (*i.e.* $S = 1/2$). For simplicity, we normalize \mathcal{I}_{GHZ} (the maximum value of N spins) to become N rather than $NS = N/2$, which can be done by multiplying 2 to the original definition of $\mathcal{I}(\rho)$.

Comparison with QFI based measure.— Although our measure and the QFI based one [21] are devised from different starting points, they have similar mathematical structures. The QFI is defined as $F(\rho, A) = 2 \sum_{i,j=1}^{2^N} (\pi_i - \pi_j)^2 / (\pi_i + \pi_j) |\langle i|A|j \rangle|^2$, where $\pi_i(|i\rangle)$ is i -th eigenvalue(eigenvector) of the density matrix ρ and $A = \sum_{j=1}^N \alpha^{(j)} \cdot \sigma^{(j)}$ with Pauli operators $\sigma^{(j)}$ for j -th site with $\|\alpha^{(j)} \cdot \sigma^{(j)}\|^2 = \|\alpha^{(j)}\|^2 = 1$. The effective size of a macroscopic quantum state is then defined as [21]

$$\mathcal{F}(\rho) \equiv \max_A \frac{F(\rho, A)}{4N} = \frac{1}{2N} \max_A \sum_{i,j=1}^{2^N} \frac{(\pi_i - \pi_j)^2}{(\pi_i + \pi_j)} |\langle i|A|j \rangle|^2, \quad (7)$$

and it has a maximum N for an N -partite GHZ state. Using Eqs. (5) and (6), we can rewrite $\mathcal{I}(\rho)$ (with an extra normalization factor 2 mentioned above) as

$$\mathcal{I}(\rho) = \frac{1}{2N} \max_A \sum_{i,j=1}^{2^N} \frac{(\pi_i - \pi_j)^2}{\sum_k \pi_k^2} |\langle i|A|j \rangle|^2. \quad (8)$$

It is clear that the only difference between $\mathcal{I}(\rho)$ and $\mathcal{F}(\rho)$ is the denominator of the weights of $|\langle i|A|j \rangle|^2$ which are $\sum_k \pi_k^2$ and $\pi_i + \pi_j$, respectively. Both $\mathcal{I}(\rho)$ and $\mathcal{F}(\rho)$

become identical to the maximum variance *per* particle, $\max_A \mathcal{V}(A)/N$, for any pure state. It was pointed out that the QFI is the convex roof of the variance, thus it is a bound for any convex function that are identical to the variance for pure states [24, 31]. However, the convexity of $\mathcal{I}(\rho)$ is not guaranteed because of the purity in the denominator in Eq. (5), and the difference between the two measures may appear for some highly mixed states as discussed in the following examples.

Examples for mixed states.— Consider a generalized mixed GHZ state

$$\rho_G = \mathcal{N}^{-1} \left(|0\rangle\langle 0|^{\otimes N} + |\epsilon\rangle\langle \epsilon|^{\otimes N} + \gamma |0\rangle\langle \epsilon|^{\otimes N} + \gamma |\epsilon\rangle\langle 0|^{\otimes N} \right) \quad (9)$$

where $|\epsilon\rangle = \cos \epsilon |0\rangle + \sin \epsilon |1\rangle$ and $\mathcal{N} = 2(1 + \gamma \cos^N \epsilon)$. The two components of the superposition $|0\rangle^{\otimes N}$ and $|\epsilon\rangle^{\otimes N}$ have an overlap $\langle 0|\epsilon\rangle^N = \cos^N \epsilon$ and their coherence is reduced by factor $\gamma \leq 1$. Using Eq. (5), we obtain $\mathcal{I}(\rho_G) \simeq 2\gamma^2/(1 + \gamma^2)\epsilon^2 N + O(1)$, where the approximation is taken only in the limit $\epsilon \ll 1$, and γ is still arbitrary. The QFI based measure is obtained as $\mathcal{F}(\rho_G) \simeq \gamma^2 \epsilon^2 N + O(1)$ by diagonalizing ρ_G with Eq. (7). The ratio between the two measure in the limit of a high mixture is $\mathcal{I}(\rho_G)/\mathcal{F}(\rho_G) \simeq 2$ ($\gamma \ll 1$ and $N \gg 1$). The two measures are identical for a pure state, $\gamma = 1$, $\mathcal{I}(\rho_G) = \mathcal{F}(\rho_G) = \max_A \mathcal{V}(A)/N \simeq \epsilon^2 N + O(1)$; this coincides with the result of an earlier study on the pure GHZ state based on the dephasing rate [12].

We investigate another type of mixed state that shows sub-optimal precision for quantum metrology [32]

$$\rho_M = \mathcal{C} H_1 \rho_0^{\otimes N} H_1 \mathcal{C} = \frac{1}{2} \begin{pmatrix} \rho_0^{\otimes N-1} & p(\rho_0 \sigma_x)^{\otimes N-1} \\ p(\sigma_x \rho_0)^{\otimes N-1} & (\sigma_x \rho_0 \sigma_x)^{\otimes N-1} \end{pmatrix}, \quad (10)$$

where $\rho_0 = \{(1+p)|0\rangle\langle 0| + (1-p)|1\rangle\langle 1|\}/2$, H_1 is the Hadamard gate on the first qubit, $\mathcal{C} = \bigotimes_{i=2}^N C_{1i}$, and C_{1i} is the controlled-NOT operation with the first qubit being the control and the i th qubit being the target. It is straightforward to obtain $\mathcal{I}(\rho_M)$ from the explicit form of Eq. (10) as $\mathcal{I}(\rho_M) = 8p^4/(1+p^2)^3 N + O(1) \simeq 8p^4 N + O(1)$ ($p \ll 1$). However, it is necessary to calculate the eigenvalues and eigenvectors of ρ_M in order to obtain its QFI, which was done in Ref. [32] and we thus get $\mathcal{F}(\rho_M) = p^4 N + O(1)$. This is a typical example that shows the computational advantage of $\mathcal{I}(\rho)$. The ratio between the two measures becomes $\mathcal{I}(\rho_M)/\mathcal{F}(\rho_M) \simeq 8$ ($p \ll 1$, $N \gg 1$) that is even larger compared to the previous example.

Quantum phase transition.— QPT is a well known quantum effect where the ground state of a many-body system shows an abrupt change of an observable called an order parameter. We investigate the transverse Ising model that is the simplest quantum many-body model exhibiting QPT [3, 33, 34]. Its Hamiltonian is

$$H_{\text{Ising}}(\lambda) = - \sum_{j=1}^N \left(\lambda \sigma_x^{(j)} \sigma_x^{(j+1)} + \sigma_z^{(j)} \right) \quad (11)$$

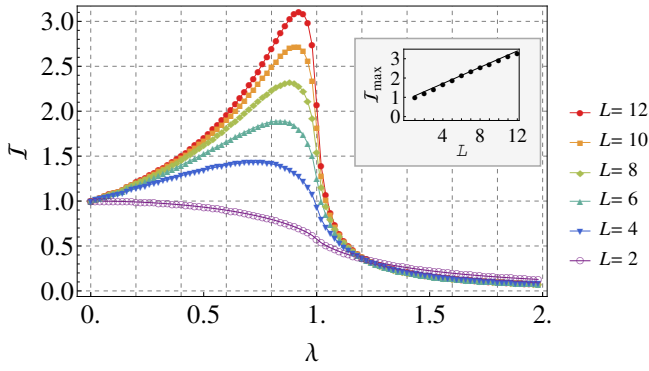


FIG. 2: Quantum macroscopicity $\mathcal{I}(\rho_L)$ of partial block of L contiguous particles in the ground state versus the interaction strength λ of the Ising model. The QPT occurs at the critical point $\lambda = 1$. (Inset) Black dots correspond to the maximal values $\mathcal{I}_{\max} = \max_{\lambda} \mathcal{I}(\rho_L(\lambda))$ for block L , and the linear fit to $\mathcal{I}_{\max} \simeq 0.21L + O(1)$.

where $\sigma_i^{(j)}$ is the i -component Pauli operator on the j -th site, λ is the interaction strength, and a periodic condition $\sigma^{(N+1)} \equiv \sigma^{(1)}$ is given. The QPT then occurs at the critical point $\lambda = \lambda_c = 1$ where the global phase-flip symmetry of the ground state is broken [33]. For $\lambda < \lambda_c$, the ground state is in disordered paramagnetic phase with the order parameter $\langle \sigma_x \rangle = 0$ while it becomes ordered ferromagnetic phase for $\lambda > \lambda_c$ with $\langle \sigma_x \rangle \neq 0$.

We take the thermodynamic limit of the model (*i.e.* $N \rightarrow \infty$) and consider a partial block of L contiguous particles of the ground state to obtain the reduced density matrix ρ_L . Note that ρ_L is generally not a pure state since L spins are originally correlated with the rest part of the spin chain. Even if we consider the ground state of the entire particles, mixedness occurs due to the two-fold degeneracy for $\lambda > 1$, which originates from the global phase-flip symmetry [33]. Such inevitable mixedness makes other existing measures, especially those only for pure states [12–14, 17, 18, 23], not applicable to this study.

In order to obtain $\mathcal{I}(\rho_L)$, we first find explicit forms of ρ_L and A in terms of the Pauli matrices for a number of given block sizes up to $L = 12$ using the analytic solution of ρ_L obtained in Ref. [3] (Supplementary Material). We put those forms into our formal definition $\mathcal{I}(\rho_L) = \max_A \{ \text{Tr}[\rho_L^2 A^2 - \rho_L A \rho_L A] \} / \{ L \text{Tr}[\rho_L^2] \}$. We then perform the numerical optimization procedure over

all possible directions of $\alpha^{(i)}$ for each site i . While we are not required to find the eigenvalues and eigenvectors of ρ_L , the numerical optimization process for L local parameters $\alpha^{(i)}$ becomes heavier as L increases because the block ρ_L has no translational symmetry due to the open boundaries.

The result is shown in Fig. 2 for block sizes up to $L = 12$. As L increases, we observe that $\mathcal{I}(\rho_L)$ peaks right before the critical point $\lambda_c = 1$ and rapidly decreases as the interaction becomes stronger. If we collect the maximum values of $\mathcal{I}(\rho_L)$ for each L , it exhibits a linear scaling as $\max_{\lambda} \mathcal{I}(\rho_L(\lambda)) \simeq 0.21L + O(1)$ as shown in the inset of Fig. 2. This implies that, for any large value of L , the system may possess a degree of quantum macroscopicity proportional to the system size at the critical point. It can be attributed to the fact that the particles acquire long-range correlations near the critical point [3, 34]. The type of correlation is quantum entanglement which has no classical counter part, and it also shows a similar scaling behavior near the critical point [34]. Our result strongly suggests that the QPT may be regarded as a genuine macroscopic quantum phenomenon where a subset of the system cannot explain the entire quantum mechanical properties.

Conclusion.— We have suggested a general and computable measure of quantum macroscopicity for arbitrary spin systems. It is based on the idea of quantifying interference fringes in the phase space and is devised to discriminate genuine macroscopic quantum effects from mere accumulations of microscopic quantum effects. It has conceptual and mathematical connections with previous measures for bosonic states [20] and spin states [21] with its own merit of the computational convenience. It enables us to explore quantum macroscopicity that arises during the QPT of many-body spin systems. Interestingly, a large degree of quantum macroscopicity emerges when the QPT occurs near the critical point. It suggests that a macroscopic quantum superposition of an extremely large size may appear during the QPT. Our study provides a strong tool to explore quantum macroscopicity of many body systems and presents a novel example where quantum macroscopicity arises during a natural many-body phenomenon.

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Supplemental Material: Measure of Quantum Macroscopicity for Arbitrary Spin Systems and Quantum Phase Transition as a Genuine Macroscopic Quantum Phenomenon

1. Purity expressed in terms of the characteristic function

An explicit form of the Wigner distribution for a spin- S particle is [S1]

$$W(\mathbf{n}) = \sqrt{\frac{4\pi}{2S+1}} \sum_{L=0}^{2S} \sum_{M=-L}^L \chi_{L,M}^{(S)} Y_{L,M}(\mathbf{n}), \quad (\text{S1})$$

where $Y_{L,M}(\mathbf{n})$ denotes spherical harmonics with a three dimensional unit vector $\mathbf{n}=(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ and $\chi_{L,M}^{(S)} = \text{Tr}[\hat{T}_{L,M}^{(S)\dagger}\rho]$ is the characteristic function with is the irreducible tensor operator $\hat{T}_{L,M}^{(S)}$ defined in the main text and the density operator ρ . The Wigner distribution can also be expressed as $\text{Tr}[\rho\hat{w}(\mathbf{n})]$ with the transformation kernel [S1]

$$\hat{w}(\mathbf{n}) = \sqrt{\frac{4\pi}{2S+1}} \sum_{L=0}^{2S} \sum_{M=-L}^L \hat{T}_{L,M}^{(S)\dagger} Y_{L,M}(\mathbf{n}). \quad (\text{S2})$$

In general, the W-symbol of an arbitrary operator \hat{f} is defined as $W_f(\mathbf{n}) = \text{Tr}[\hat{f}\hat{w}(\mathbf{n})]$ and the expectation value of \hat{f} can be obtained by

$$\text{Tr}[\rho\hat{f}] = \frac{2S+1}{4\pi} \int d\Omega W(\mathbf{n})W_f(\mathbf{n}). \quad (\text{S3})$$

Using the orthogonality of the spherical harmonics,

$$\int d\Omega Y_{L,M}(\mathbf{n})Y_{L',M'}^*(\mathbf{n}) = \delta_{L,L'}\delta_{M,M'}, \quad (\text{S4})$$

we can inversely obtain the characteristic function from the Wigner distribution as

$$\begin{aligned} & \sqrt{\frac{2S+1}{4\pi}} \int d\Omega Y_{L,M}^*(\mathbf{n})W(\mathbf{n}) \\ &= \sum_{L',M'} \int d\Omega Y_{L',M'}(\mathbf{n})Y_{L,M}^*(\mathbf{n})\chi_{L',M'}^{(S)} \\ &= \sum_{L',M'} \delta_{L,L'}\delta_{M,M'}\chi_{L',M'}^{(S)} \\ &= \chi_{L,M}^{(S)}. \end{aligned} \quad (\text{S5})$$

We then find a relation between the purity $\mathcal{P}(\rho)$ of state ρ and the characteristic function $\chi_{L,M}^{(S)}$ as

$$\begin{aligned} & \sum_{L,M} |\chi_{L,M}^{(S)}|^2 \\ &= \frac{2S+1}{4\pi} \int d\Omega d\Omega' \left(\sum_{L,M} Y_{L,M}(\mathbf{n})Y_{L,M}^*(\mathbf{n}') \right) W(\mathbf{n})W(\mathbf{n}') \\ &= \frac{2S+1}{4\pi} \int d\Omega d\Omega' \delta(\mathbf{n}-\mathbf{n}') W(\mathbf{n})W(\mathbf{n}') \\ &= \frac{2S+1}{4\pi} \int d\Omega W^2(\mathbf{n}) = \text{Tr}[\rho^2] = \mathcal{P}(\rho). \end{aligned} \quad (\text{S6})$$

2. Wigner representation of the measure of quantum macroscopicity

We can prove Eq. (2) in the main text as follow. Using Eq. (S5) in the previous section and the theorem of

integration by parts, we find

$$\begin{aligned}
\mathcal{I}_z &= \frac{1}{2S\mathcal{P}} \sum_{L=0}^{2S} \sum_{M=-L}^L M^2 \left| \chi_{L,M}^{(S)} \right|^2 \\
&= \frac{1}{2S\mathcal{P}} \sum_{L=0}^{2S} \sum_{M=-L}^L M^2 \chi_{L,M}^{(S)*} \chi_{L,M}^{(S)} \\
&= \frac{1}{2S\mathcal{P}} \sqrt{\frac{2S+1}{4\pi}} \sum_{L,M} \int d\Omega M^2 Y_{L,M}(\mathbf{n}) W(\mathbf{n}) \chi_{L,M}^{(S)} \\
&= -\frac{1}{2S\mathcal{P}} \sqrt{\frac{2S+1}{4\pi}} \sum_{L,M} \int d\Omega \frac{\partial^2 Y_{L,M}(\mathbf{n})}{\partial \phi^2} W(\mathbf{n}) \chi_{L,M}^{(S)} \\
&= -\frac{2S+1}{8\pi S\mathcal{P}} \sum_{L,M} \int d\Omega d\Omega' Y_{L,M}(\mathbf{n}) \frac{\partial^2 W(\mathbf{n})}{\partial \phi^2} Y_{L,M}^*(\mathbf{n}') W(\mathbf{n}') \\
&= -\frac{2S+1}{8\pi S\mathcal{P}} \int d\Omega d\Omega' \delta(\mathbf{n} - \mathbf{n}') \frac{\partial^2 W(\mathbf{n})}{\partial \phi^2} W(\mathbf{n}') \\
&= -\frac{2S+1}{8\pi S\mathcal{P}} \int d\Omega \frac{\partial^2 W(\mathbf{n})}{\partial \phi^2} W(\mathbf{n}) \\
&= \frac{2S+1}{8\pi S\mathcal{P}} \int d\Omega W(\mathbf{n}) \hat{L}_z^2 W(\mathbf{n}), \tag{S7}
\end{aligned}$$

so that the two lines of Eq. (2) in the main text are identical.

3. Density operator representation of the measure of quantum macroscopicity

We now show that the two definitions of \mathcal{I}_α , Eq. (3) of the main text and the objective function that undergoes the maximization in Eq. (5) of the main text, for a single particle are equivalent. The density matrix of \mathcal{I}_α can be expressed as

$$\begin{aligned}
\mathcal{I}_\alpha &= \frac{1}{S\mathcal{P}} (\text{Tr} [\rho^2 A^2] - \text{Tr} [\rho A \rho A]) \\
&= \frac{1}{S\mathcal{P}} \sum_{i,j=x,y,z} \alpha_i \alpha_j (\text{Tr} [\rho^2 S_i S_j] - \text{Tr} [\rho S_i \rho S_j]) \\
&= \frac{1}{S\mathcal{P}} \sum_{i,j=x,y,z} \alpha_i \alpha_j \text{Tr} [\rho \hat{f}_{ij}], \tag{S8}
\end{aligned}$$

where $\hat{f}_{ij} = ([S_i, S_j \rho] + [\rho S_i, S_j])/2$. Since the trace of any two operators can be calculated as Eq. (S3), we need to obtain the W-symbol of \hat{f}_{ij} as

$$\begin{aligned}
W_{f_{ij}} &= \text{Tr} [\hat{f}_{ij} \hat{w}(\mathbf{n})] = \frac{1}{2} \text{Tr} [S_i S_j \rho \hat{w} + \rho S_i S_j \hat{w} - 2 S_j \rho S_i \hat{w}] \\
&= \frac{1}{2} \text{Tr} [\rho \hat{w} S_i S_j + \rho S_i S_j \hat{w} - 2 \rho S_i \hat{w} S_j] \\
&= \frac{1}{2} \text{Tr} [\rho ([\hat{w}, S_i] S_j + S_i [S_j, \hat{w}])] \\
&= \frac{1}{2} \hat{L}_i \text{Tr} [\rho \hat{w} S_j] - \frac{1}{2} \hat{L}_j \text{Tr} [\rho S_i \hat{w}], \tag{S9}
\end{aligned}$$

where the last equality holds for $[\hat{w}, S_i] = \hat{L}_i \hat{w}$, which is proven in Ref. [S2]. When $i = j$, it is further simplified as

$$W_{f_{ii}} = \frac{1}{2} \hat{L}_i \text{Tr} [\rho [\hat{w}, S_i]] = \frac{1}{2} \hat{L}_i^2 \text{Tr} [\rho \hat{w}] = \frac{1}{2} \hat{L}_i^2 W(\mathbf{n}). \tag{S10}$$

If $i \neq j$, \hat{f}_{ij} always has its symmetric pair \hat{f}_{ji} in Eq. (S8) so that we can combine them as

$$\begin{aligned}
W_{f_{ij}} + W_{f_{ji}} &= \text{Tr} [(\hat{f}_{ij} + \hat{f}_{ji}) \hat{w}] \\
&= \frac{1}{2} \hat{L}_i \text{Tr} [\rho [\hat{w}, S_j]] + \frac{1}{2} \hat{L}_j \text{Tr} [\rho [\hat{w}, S_i]] \\
&= \frac{1}{2} (\hat{L}_i \hat{L}_j + \hat{L}_j \hat{L}_i) \text{Tr} [\rho \hat{w}] \\
&= \frac{1}{2} (\hat{L}_i \hat{L}_j + \hat{L}_j \hat{L}_i) W(\mathbf{n}). \tag{S11}
\end{aligned}$$

Using Eqs. (S3), (S10) and (S11), Eq. (S8) becomes

$$\begin{aligned}
\mathcal{I}_\alpha &= \frac{2S+1}{8\pi S\mathcal{P}} \sum_{i,j=x,y,z} \alpha_i \alpha_j \int d\Omega W(\mathbf{n}) \hat{L}_i \hat{L}_j W(\mathbf{n}) \\
&= \frac{2S+1}{8\pi S\mathcal{P}} \int d\Omega W(\mathbf{n}) \hat{L}_\alpha^2 W(\mathbf{n}). \tag{S12}
\end{aligned}$$

Its extension to an arbitrary number of spin is straightforward because the measure contains at most the quadratic order of spin operators and the operators for different sites commute.

4. Upper bound of the measure of quantum macroscopicity

We here show that the upper bound of $\mathcal{I}(\rho)$ for an arbitrary multipartite system ρ composed of N spin- S particles is NS . We start from the definition

$$\mathcal{I}(\rho) = \frac{1}{NS\mathcal{P}} \max_A \text{Tr} [\rho^2 A^2 - \rho A \rho A] \tag{S13}$$

in Eq. (5) of the main text. By noting that $\text{Tr}[(\rho A)^2] \geq 0$, we find

$$\begin{aligned}
&\text{Tr} [\rho^2 A^2] - \text{Tr} [\rho A \rho A] \\
&\leq \text{Tr} [\rho^2 A^2] = \sum_{i=1}^{2^N} \pi_i^2 \langle i | A^2 | i \rangle = \sum_{i=1}^{2^N} \pi_i^2 \sum_{j,k=1}^N \langle i | A^{(j)} A^{(k)} | i \rangle \\
&\leq \sum_{i=1}^{2^N} \pi_i^2 \sum_{j,k=1}^N S^2 = \sum_{i=1}^{2^N} \pi_i^2 N^2 S^2 = \mathcal{P} N^2 S^2, \tag{S14}
\end{aligned}$$

where $\pi_i (|i\rangle)$ is the i -th eigenvalue (eigenvector) of the density operator ρ . Using Eqs. (S13) and (S14), we obtain

$$\mathcal{I}(\rho) \leq \frac{1}{NS\mathcal{P}} \mathcal{P} N^2 S^2 = NS. \tag{S15}$$

As an example, we find that the N -partite spin- S GHZ state,

$$|\text{GHZ}\rangle = \frac{1}{\sqrt{2}} (|S, S\rangle^{\otimes N} + |S, -S\rangle^{\otimes N}), \quad (\text{S16})$$

has the maximal value $\mathcal{I}(\rho) = NS$ when the operator $A^{(i)}$ is aligned along the z -direction for every value of i .

5. Calculation of \mathcal{I} for the ground state of Ising model

The Hamiltonian of the Ising model is given by

$$H_{\text{Ising}}(\lambda) = - \sum_{j=1}^N \left(\lambda \sigma_x^{(j)} \sigma_x^{(j+1)} + \sigma_z^{(j)} \right), \quad (\text{S17})$$

where $\sigma_i^{(j)}$ is the i -component Pauli operator on the j th site, λ is the interaction strength, and a periodic condition $\sigma^{(N+1)} \equiv \sigma^{(1)}$ is given. We here briefly review an analytic method to obtain the ground state of this model [S3] and calculate \mathcal{I} for it. The Hamiltonian in Eq. (S17) can be diagonalized using two Majorana operators for each site l of the N spins,

$$c_{2l} \equiv \left(\prod_{m=0}^{l-1} \sigma_m^z \right) \sigma_l^x \quad \text{and} \quad c_{2l+1} \equiv \left(\prod_{m=0}^{l-1} \sigma_m^z \right) \sigma_l^y, \quad (\text{S18})$$

where c_m is a Hermitian operator and satisfies the anti-commutation relation, $\{c_m, c_n\} = 2\delta_{mn}$. It is known that the expectation values $\langle c_m c_n \rangle = \delta_{mn} + i\Gamma_{mn}$ completely characterize L block of the ground state [S3], where the matrix Γ is has the form,

$$\Gamma = \begin{bmatrix} \Pi_0 & \Pi_1 & \cdots & \Pi_{L-1} \\ \Pi_{-1} & \Pi_0 & & \vdots \\ \vdots & & \ddots & \vdots \\ \Pi_{1-L} & \cdots & \cdots & \Pi_0 \end{bmatrix}, \quad \Pi_l = \begin{bmatrix} 0 & g_l \\ -g_{-l} & 0 \end{bmatrix}, \quad (\text{S19})$$

where g_l is given by

$$g_l = \frac{1}{2\pi} \int_0^{2\pi} d\phi e^{-il\phi} \frac{\lambda e^{-i\phi} - 1}{|\lambda e^{-i\phi} - 1|}. \quad (\text{S20})$$

Since Γ is a skew-symmetric matrix, we can find an orthogonal matrix $V \in \text{SO}(2L)$ that block-diagonalizes Γ

into

$$\tilde{\Gamma} = V\Gamma V^T = \oplus_{m=0}^{L-1} \nu_m \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}. \quad (\text{S21})$$

The set of $2L$ Majorana operators $d_m = \sum_{n=0}^{2L-1} V_{mn} c_n$ have a block-diagonal correlation matrix $\langle d_m d_n \rangle = \delta_{mn} + i\tilde{\Gamma}_{mn}$. Now, L fermionic operators $b_l \equiv (d_{2l} + id_{2l+1})/2$ obeying $\{b_m, b_n\} = 0$ and $\{b_m^\dagger, b_n\} = \delta_{mn}$ have expectation values

$$\langle b_m \rangle = 0, \quad \langle b_m b_n \rangle = 0, \quad \langle b_m^\dagger b_n \rangle = \delta_{mn} \frac{1 + \nu_m}{2}. \quad (\text{S22})$$

This indicates that the mixed states of the block L can be described as a product state in b_m basis as

$$\rho_L = \bigotimes_{m=0}^{L-1} \rho_m. \quad (\text{S23})$$

We re-express Eq. (S23) in the Pauli operator basis. From Eq. (S22), one can find

$$\rho_m = \frac{1 - \nu_m}{2} b_m b_m^\dagger + \frac{1 + \nu_m}{2} b_m^\dagger b_m, \quad (\text{S24})$$

and we can expand b_m as

$$\begin{aligned} b_m &= \frac{1}{2} (d_{2l} + id_{2l+1}) = \frac{1}{2} \sum_{n=0}^{2L-1} (V_{2l,n} c_n + iV_{2l+1,n} c_n) \\ &= \frac{1}{2} \left[\sum_{k=0}^{L-1} (V_{2l,2k} + iV_{2l+1,2k}) c_{2k} \right. \\ &\quad \left. + \sum_{k=0}^{L-1} (V_{2l,2k+1} + iV_{2l+1,2k+1}) c_{2k+1} \right] \\ &= \frac{1}{2} \left[\sum_{k=0}^{L-1} (V_{2l,2k} + iV_{2l+1,2k}) \left(\prod_{m=0}^{k-1} \sigma_m^z \right) \sigma_l^x \right. \\ &\quad \left. + \sum_{k=0}^{L-1} (V_{2l,2k+1} + iV_{2l+1,2k+1}) \left(\prod_{m=0}^{k-1} \sigma_m^z \right) \sigma_l^y \right]. \end{aligned} \quad (\text{S25})$$

We now have an analytic form of $\mathcal{I}(\rho_L)$ since ρ_L and $A = \sum_{j=1}^N \alpha^{(j)} \cdot \sigma^{(j)}$ are all written in the Pauli basis. However, the measure requires maximization over the local unit vectors $\alpha^{(j)}$ s of the operator A . Since this multi-dimensional optimization cannot be done analytically, we numerically obtain the final values of $\mathcal{I}(\rho_L)$ using the steepest decent method [S4].

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